

BIOTRANSFORMATION OF METAL CONTAMINANTS IN SOILS/SEDIMENTS: CHROMIUM

Tetsu Tokunaga, Jiamin Wan, Egbert Schwartz,¹ Dominique Joyner, Stephen Sutton,² Matt Newville,² Mary Firestone¹ and Terry Hazen

¹UC Berkeley; ²University of Chicago

Contact: Tetsu Tokunaga, 510/486-7176, tktokunaga@lbl.gov

RESEARCH OBJECTIVES

Understanding transport and reactions of metal contaminants such as chromium in soils is complicated by small-scale variations in physical, chemical and microbiological characteristics. The fate of elements with highly redox dependent solubilities can be especially complex because strong redox potential gradients can develop over very short distances. In nature, Cr exists in the III and VI oxidation states, with the majority of the former species being stable solids, and the majority of the latter being more soluble and mobile. Transport and reactions of chromium in soils are critical concerns because of the carcinogenic effects of Cr(VI). Cr reduction rates reflect interdependent influences of physical, geochemical and microbial processes. The overall process tested in this study is the response of soil aggregates (cohesive structural units comprised of many primary mineral particles) to a Cr(VI) contamination event.

APPROACH

The interdependent influences that sediment structure and microbial communities have on transport and reduction of chromate are being investigated in soil aggregate (Altamont clay) microcosms. These microcosms were typically 30 mm in length, with an aerobic boundary at one end, and a potentially anaerobic core region on the opposite end. The soils were saturated with solutions containing 0 to 800 ppm organic carbon (OC). More rapid microbial growth, hence more reducing conditions, result from OC amendments. Following 14 days of incubation, the exterior boundary of each microcosm was exposed with up to 5200 ppm Cr(VI) solution for three days, representing a contamination event. Micro-XANES (x-ray absorption near edge structure) spectroscopy was used because of interest in Cr oxidation state determination, and the need for spatially-resolved measurements. Micro-XANES profiles of Cr(VI) and Cr(III) were measured at the GSECARS microprobe beamline (Advanced Photon Source, Argonne National Laboratory). Aggregates were later sectioned for characterization of microbial population profiles and other chemical analyses.

ACCOMPLISHMENTS

Redox measurements in the sediment microcosms showed lower potentials in systems with higher OC addition, and more oxidizing conditions within 2 to 4 mm of aggregate surfaces. Micro-XANES spectroscopy showed short Cr penetration distances, with abrupt rather than diffuse termination, and direct evidence of nearly complete Cr(VI) reduction to Cr(III). The extent of Cr transport into sediment blocks was far less than expected by diffusion without reduction, inversely related to OC amendment, and proportional to the boundary Cr(VI) concentration. Microbial communities were characterized with DNA fingerprints, direct counting and enrichment culturing. The microbial community composition in the exposure region is different from those in sediments taken from greater depth. Several populations appear only in soil that was exposed to Cr, suggesting that they are chromium resistant and that they may play an active role in Cr reduction.

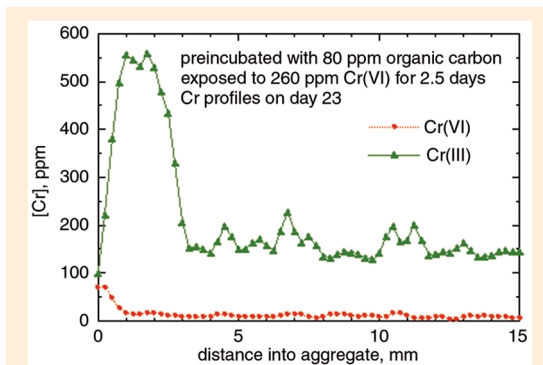


Figure 1. Profiles of Cr(VI) and Cr(III) in a soil aggregate, 23 days after exposure to Cr(VI) at the surface ($x = 0$ mm). The aggregate surface was in contact with a 260-ppm Cr(VI) solution for 2.5 days.

Direct counting of microbial populations in the sediment microcosms showed higher population densities in the outer layers due to the availability of oxygen.

SIGNIFICANCE OF FINDINGS

These results show how a metal contaminant, Cr(VI), can become very locally reduced within soil aggregates. In systems with high microbial activity, reducing conditions can develop at the mm scale. The spatial resolution needed for understanding such systems is determined by characteristic transport distances, and these distances are determined by diffusivities and reaction times. The need for measurements with at least mm-scale spatial resolution was demonstrated for highly nonequilibrium reactive transport of Cr within soil aggregates. In such systems, coarser scale volume-averaged chemical speciation will not permit mechanistic characterization of reactive transport.

ACKNOWLEDGEMENTS

We thank Keith Olson, Andrew Mei and H. Scott Mountford of LBNL, and GSECARS staff for assistance. This work has been supported by the Office of Science, Office of Basic Energy Sciences, Geosciences Research Program, and NABIR program, of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098.